

# On the preparation of radioactive samples by means of electromagnetic isotope separation

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With 9 figures in the text

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## ABSTRACT

A small electromagnetic isotope separator has been constructed with special regard to investigations of synchrocyclotron-produced radioactivities. The requirements put forward, including the simultaneous collection at equal focusing conditions of a large fraction of the mass spectrum, were found to be fulfilled by the parallel beam type of magnetic analyser. A brief review of the basic principles of the apparatus is given. Various measures aiming at increased efficiency of the process, improved focusing at higher ion currents, and more stable running conditions are discussed.

The performance of the separator is illustrated by some actual investigations. The applications include mass assignments,  $\beta$ - and  $\gamma$ -ray spectrometry, half-life determinations in the region below 1 ms, measurements of relative yields of spallation and high energy fission products, and "hot atom chemistry".

## 1. Introduction

A considerable part of the radioactive nuclei which are of current interest to nuclear research can be obtained only by nuclear reactions where, even with anisotropic target material, more than one product nuclide is formed. This means that a detailed study of their individual properties may meet with difficulties, if it is not rendered impossible, when merely a chemical separation of the reaction products is performed. In this situation the application of mass-spectroscopic methods offers an obvious way of solving the problems.

The ion currents necessary to make sufficiently rapid separations (down to some ten minutes' duration) of useful amounts of radioactive material are in principle very modest. This can be readily seen by using the Faraday law, the Avogadro number, and fundamental definitions of radioactivity to derive a production formula:

$$\Lambda_{[\text{mC}]} = 1.2 \cdot 10^5 \cdot T_{[\text{hr}]} \cdot i_{[\mu\text{A}]} \cdot \tau_{\frac{1}{2}}^{-1} \quad (1)$$

which is valid for singly charged ions. Thus in a separation time  $T$  equal to the half-life  $\tau_{\frac{1}{2}}$ , an amount corresponding to about 1 mC is transported by the ion current  $10^{-5} \mu\text{A}$ .<sup>1</sup> This is well within the intensity region of for instance a Nier type mass spectrometer.

It is reasonable to ask, then, why such simple and commercially available apparatus has not been extensively used in preparing radioactive samples. Several reasons may be suggested, the most important being the low overall efficiency, causing serious problems as to the initial activities demanded, and the unfeasibility in many cases of introducing the active material carrierfree into the ion source, which means that the much stronger currents due to ions of stable nuclides will overload the apparatus.

Isotope separators of the calutron type [1, 2, 3] on the other hand, which can handle considerable ion currents (several hundred mA) and have much more efficient ion sources, are not convenient for use in ordinary research laboratories. Besides they are constructed to give enrichment rather than the almost complete separation that is necessary in many radioactive applications.

Consequently, a flexible instrument is desirable, combining the good resolving power of the mass spectrometer with the high efficiency of isotope separators and capable of working with intermediate ion currents. In Denmark and Sweden a development along these lines has been going on for several years, originating in the application of Koch's high intensity mass spectrograph [4] to the separation of radioactive krypton isotopes produced in fission [5]. The potentialities of this technique in nuclear spectroscopy were beautifully demonstrated by Bergström [6] and Thulin [7], who used the Stockholm separator [8] in extensive investigations of Kr and Xe isotopes and even in work with solid charge material.

At this institute a small isotope separator (usually referred to as "the mass separator") has been in operation for about four years. Major attention has been paid to the separation, for various uses, of the products from high energy reactions in our 190 MeV synchrocyclotron. The requirements imposed on the separation technique by this field of application are discussed in section 2 of this paper. Though the separa-

<sup>1</sup> It should be noted that only in the limit  $T/\tau_{\frac{1}{2}} \rightarrow 0$  does  $\Lambda$  represent the activity present at the collector after the separation (assuming 100 % collection efficiency).

tor concerned has several features in common with those mentioned above and with one built in Gothenburg [9], it is thought that the differences are essential enough to warrant the description of the apparatus given in section 3. Section 4 gives an account of a number of actual investigations, and in the concluding remarks (section 5) future development and possible improvements are discussed on the basis of the experience gained.

## 2. Method

### 2.1. Radioisotope separation versus stable isotope separation

When half-lives below a few hours are concerned, special problems which are not met with in stable isotope separation become important. While for instance in the calutrons a start-up period of several hours may be allowed in order to reach steady conditions of operation, a radioactive separation often has to begin a few minutes after the introduction of the charge material. This requires rapid vacuum gating and evacuation to appropriate pressure. The construction of the ion source is also greatly influenced, a low heat capacity and good thermal insulation being imperative for the evaporation oven and the discharge chamber. It is still very likely that the overall efficiency has to be sacrificed in comparison to the stable case. This certainly constitutes a difficult matter of compromise.

In rare cases a cyclotron target can be subjected to mass separation directly after irradiation, but as a rule there must be a chemical procedure in between. Then time does not always allow the most suitable charge material for the separator to be prepared. Again the separation efficiency and the time factor oppose each other.

The collection of intense ion beams is known to constitute a serious problem in stable isotope separation because of reflection and sputtering effects [10]. With the low intensities used in the radioactive separations, however, the method of direct deposition [11] may be generally applied. In this way the samples can be brought to measurement without subsequent chemical procedures, which in fact is one of the main advantages of the technique and fundamental for its application to reaction yield studies (cf. section 2.3.). The mechanism of ion collection with special regard to direct deposition has been further studied by Thulin [12] and Nielsen [13].

An admixture of stable isotopes in the charge material is, however, convenient for several reasons. It facilitates a rapid chemistry, it makes possible the use of more efficient ion sources, and finally, it furnishes ion beams of sufficient strength to be visually controlled when impinging on a fluorescent screen or merely a glass plate and to be used in stabilizing the position of the mass spectrum (section 3.3). The amount to be added varies, but a consideration of actual applications led to the conclusion that the separator should be constructed to handle total ion currents of up to a few hundred  $\mu\text{A}$ .

### 2.2. General properties of the samples

Samples for particle counting obtained by direct deposition in the mass separator are advantageous in many respects. This has been pointed out in several connections [6, 12, 14, 15] and here only a brief summary will be given.

The mass assignment is certain, provided that the resolving power of the separator is sufficient. Except for nuclides having a stable isomer the samples are in principle carrier-free. Thin backings (i.e.  $150 \mu\text{g}/\text{cm}^2$  Al foils or still thinner organic films) may be used in order to reduce back-scattering. The kinetic energy of the ions (50–70

keV) enables them to penetrate and be trapped in the collector material, that is, no additional covers are needed even with highly volatile and non-reactive substances such as the noble gases.

The penetration in itself, however, corresponds to covering the sample with an absorber, which in actual cases may amount to  $1\text{--}10\text{ }\mu\text{g}/\text{cm}^2$  as calculated from a formula given by Nielsen [13]. This is no doubt a source of distortion in extremely low energy  $\beta$ -spectroscopy. A reduction of the accelerating voltage leads to an improvement, at the cost of the resolving power, however (cf. section 3.2). It should in principle be possible to solve the problem by decelerating the ions immediately before they reach the collector, but this may meet with practical difficulties [cf. 16].

### 2.3. *Special requirements*

Two types of application have in particular influenced the construction and further development of the mass separator:  $\beta$ -spectroscopy and the study of fission and spallation yields. The separation technique is essentially the same in both cases; some differences will be made clear in the course of the discussion.

Instruments for radioactivity measurements are designed for more or less definite dimensions of the source. To meet such requirements with the method of direct deposition it is of course possible to place a defining slit in front of the collector during the mass separation. In order not to loose too much activity on the edges of these slits, however, the cross section of the beam should as far as possible have the same dimensions. The determining factor as to the desired shape and size of the deposits was in our case the  $\beta$ -spectrometer of the institute [17], which is of the two-directional focusing type. A source height of about 20 mm is suitable and in high resolution work the width should preferably be less than 1 mm.

In yield studies the separator is used to prepare samples for  $\beta$ -counting, normally performed with end-window G.M. tubes. Point sources would be ideal, but a different shape is acceptable, provided it is the same for all mass numbers concerned. In this case slits should be avoided.

Another important aspect of yield measurements is the necessity of simultaneous collection of several (sometimes  $>10$ ) isotopes of an element. Thus a large relative mass difference must be covered, with due regard to the requirement of equal focusing conditions for all the masses. This is certainly not unimportant even in the preparation of  $\beta$ -spectrometer samples if the isotope of interest is situated far away from the stable one used as a "mass marker" (section 2.1.).

## 3. Apparatus

### 3.1. *Principles of the magnetic analyser*

The lens and prism actions of magnetic fields on charged particles moving in a perfect vacuum have been treated extensively by several authors [e. g. 18, 19, 20]. The present discussion concerns the especially simple case visualized in fig. 1.

A parallel, monoenergetic ion beam of circular cross section (diameter  $d$ ) enters a homogeneous magnetic field at right angles to the plane field boundary, idealized as a discontinuous change. After  $90^\circ$  deflection the ions are brought to discrete foci according to mass. In a direction along the field lines the dimension of the beam is retained. Thus the different masses present give rise to "lines" of height  $d$  and width  $B$  (because of imperfect focusing, see below) spaced a distance  $D$  apart in a plane at  $45^\circ$  to the entrance direction.

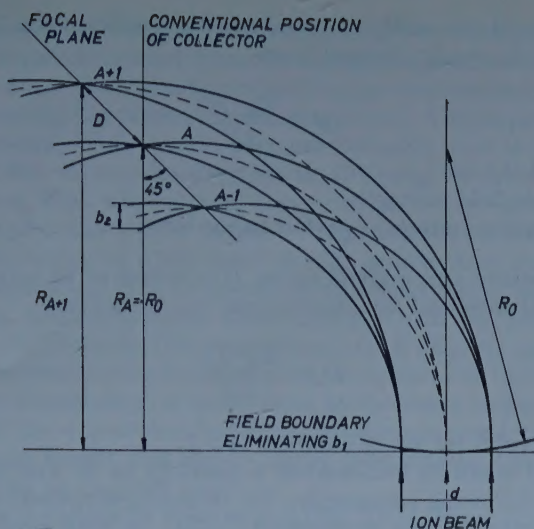


Fig. 1. Focusing of a parallel ion beam in a homogeneous magnetic field.

It should at once be remarked that the plane field boundary gives a contribution to the line width  $B$  which can be calculated using simple geometry as

$$b_1 \approx \frac{d^2 \sqrt{2}}{8R}; \quad \left( \frac{d^2}{4R^2} \text{ assumed } < 1 \right). \quad (2)$$

where  $R$  is the radius of curvature of the ions. A focusing of higher order can be achieved by making the entrance boundary convex with the same radius of curvature. If this is chosen equal to the mean radius  $R_0$ , the remaining error for  $R \neq R_0$  will be negligible with reasonable values of  $d$  and  $R$ .

The necessity in our case of equal focusing conditions for as large part as possible of the mass spectrum was pointed out in section 2. It is directly seen from the exaggerated scale in fig. 1 that this is not compatible with a "conventional" position of the collectors, that is perpendicular to the outgoing beams. Optimal line sharpness is then obtained only for one single mass, corresponding to radius  $R_0$ . For another radius  $R$  the line width contribution  $b_2$  is given by

$$b_2 \approx \frac{d \cdot \Delta R}{R}; \quad \left( \left[ \frac{2\Delta R + d}{2R} \right]^2 \text{ assumed } < 1 \right). \quad (3)$$

where  $\Delta R = |R - R_0|$ . By placing the collectors along the focal plane this error is eliminated.

### 3.2. Resolving power

The condition for complete separation of neighbouring lines in the spectrum is evidently that

$$D/B > 1. \quad (4)$$

$D$ , the dispersion of the analyser, can be calculated from the "mass-spectrometer equation", applied to singly charged ions:

$$M = \frac{R_M^2 \cdot H^2}{2V}, \quad (5)$$

where  $M$  = mass of the ion

$H$  = magnetic field strength

$V$  = acceleration voltage imparted to the ions.

For a given geometry and a given mass,  $D$  is found to be a function of  $R$  alone, which in the actual case and for atomic ions can be written:

$$D_A \approx \frac{R_A \cdot \sqrt{2}}{2A}. \quad (6)$$

This expression is valid for mass numbers  $A \geq 1$ .

The line width  $B$  is not as well-defined a quantity as  $D$ . For reasons that will be apparent from the following paragraphs, the intensity distribution across a line has a bell-shape, with more or less pronounced "tails" on both sides of a central core. Depending on the amounts of contamination that can be accepted  $B$  may be defined in different ways, for instance to include a certain percentage (99 % or 99.9 % etc.) of the total intensity of the mass number concerned. In calculating the resolving power  $P$  the width of the lines at half the maximum intensity,  $B_{\frac{1}{2}}$ , is often used:

$$P = \frac{D_A \cdot A}{B_{\frac{1}{2}}}. \quad (7)$$

In section 3.1 several idealizations were accepted which do not hold good in practice. Energy inhomogeneity and mutual repulsion of the ions, scattering against residual gas molecules, and aberrations of the magnetic analyzer may give significant contributions to  $B$ . Some of the effects can easily be treated quantitatively on the basis of Eq. 5. The energy spread in the ion source and variations in the accelerating voltage lead to terms of the type

$$b_3 \approx \frac{R\sqrt{2}}{2} \cdot \frac{\Delta V}{V}; \quad (\Delta V \text{ assumed } \ll V), \quad (8)$$

while time variations of the magnetic field give rise to the error

$$b_4 \approx R\sqrt{2} \cdot \frac{\Delta H}{H}; \quad (\Delta H \text{ assumed } \ll H). \quad (9)$$

In this way requirements are imposed on the performance of the ion source and on the stabilization of high tension and magnet current.

Eq. 8 indicates the advantage of a high acceleration voltage. This is also effective in reducing the influence of mutual repulsion (space charge broadening) as seen from the expression [4, 21]

$$\frac{d}{d_0} \approx \text{const.} \cdot \frac{A^{1/4} \cdot j_0^{3/2}}{V^{3/4}} \cdot z; \quad \left(2 < \frac{d}{d_0} < 5\right), \quad (10)$$

where  $d_0$  is the original diameter of a parallel beam,  $j_0$  the corresponding ion current density, and  $z$  the distance traversed when the diameter has increased to  $d$ . In practice secondary electrons from the ionization of residual gas molecules play a most important rôle in compensating the positive space charge [22]. This is particularly so in the high intensity separators. With the ion currents normally used in the actual type of apparatus, however, the current densities are small except in the region immediately outside the ion source. This will be more closely examined in section 3.4.

A good vacuum and a short path length of the ions are essential in keeping the spread due to gas-scattering at a low level. A high acceleration voltage seems to be favourable in this respect too [23]. It should be especially noted that all the terms of  $B$  mentioned in this section increase about linearly with the path length, that is approximately with  $R$ . Thus, as  $D$  is proportional to  $R$  (Eq. 6), nothing is gained in resolving power by increasing the dimensions of the apparatus, under the assumption that these terms are dominating. A lower limit to the mean radius  $R_0$  is put by the dispersion necessary for a practical collector arrangement and to some extent by the ratio between gap height and radius (stray field effects) [23]. We chose  $R_0 = 100$  cm, corresponding to  $D_{240} \approx 3$  mm.

In this connection it should be pointed out that  $D/L$ , where  $L$  is the path length, rather than  $D/R$  [cf. 24] is a suitable "figure of merit" in comparing different types of separators.

### 3.3. Line positioning

Even if the magnet current and the accelerating voltage are well stabilized against mains voltage fluctuations, changes in the position of the mass spectrum may occur, caused for instance by temperature effects in the magnet or electrostatic charging with subsequent sparking in the lens system. This source of contamination can be counteracted by letting a reference line of sufficient strength fall between two thin metal pins ("sniffers"), insulated from each other and forming parts of a bridge circuit. Any unbalance caused by a movement of the line is amplified and fed back to influence some parameter determining the position of the spectrum. The distance between the pins should be chosen so that they cut the sides of the reference line where the intensity gradient is maximal.

Thulin [12] has described a system where the angle of entrance of the beam into the magnetic field is varied by means of a pair of electrostatic deflector plates acted upon by the unbalance in the bridge circuit. A change of the entrance angle, however, has obviously a defocusing effect. It can further be shown that in principle only the reference line is strictly kept in position. Though both these errors are normally small with the deflection angles needed in practice, at least the latter receives increased importance when masses far away from the reference line are to be collected. Another drawback which is serious in our case is the reduction of the number of masses possible to focus simultaneously, the useful width of the magnetic field being given. Finally it should be remarked that unaltered ion trajectories are imperative for space charge compensation by secondary electrons because of the finite time taken to build up a charge equilibrium [25]. Thus additional defocusing is to be expected.

Preliminary experiments with this type of positioner seemed to confirm the apprehensions and led us to choose a different principle. The basic idea is to let the signal from the "sniffers" operate on the magnet current stabilizer. This eliminates all the troubles mentioned in the preceding paragraph and would be the ideal solution,

were it not for the long time constant of the magnet coil circuit, making the neutralization of rapid changes impossible. Therefore, the high frequency part of the signal is selected and fed to deflector plates at the magnet entrance. The magnetic field "takes over" the stabilization little by little and so the deviations from perpendicular entrance are of short duration. Further the angle of deflection is limited, an additional pair of plates bringing about a vertical deflection of the spectrum, that is, away from the collector, when the preset limit is reached. The destruction of the space charge compensation still remains, however. It would in principle be avoided if the regulation were applied to the accelerating voltage, but this on the other hand may lead to defocusing because of changed conditions of beam extraction from the ion source.

### 3.4. *Formation of the beam*

A question of major importance to the realization in practice of the focusing case described above still remains to be discussed: the possibility of forming a sufficiently parallel ion beam.

In principle a divergent beam of charged particles (from a virtual point source) can be rendered parallel by an electrostatic lens system, consisting of three cylindrical electrodes. The following example of the potential distribution is taken from Koch [26]: ion source  $+45$  kV ( $=V_A$ ), 1st lens electrode grounded, 2nd electrode (focusing electrode)  $+30$  kV ( $=V_F$ ), and 3rd electrode grounded. Koch found, however, that the expected focusing conditions were not fulfilled with this arrangement; the most favourable  $D/B$  ratio was in fact obtained at a distance equal to the radius away from the magnet edge [4].

In their separator, Bergström *et al.* [8] introduced an additional "suction electrode", kept some  $10$  kV ( $=V_s$ ) negative in relation to the ion source and placed between this and the first grounded electrode.  $V_s$  was mainly meant to determine the magnification of the system and thus to make it possible to work with fairly constant ion current density for different values of the total ion current, while still collecting the ions immediately at the magnet edge. This Stockholm version of the lens system was accepted for our mass separator.

Early experiments showed that satisfactory focusing could be obtained, provided the ion currents were sufficiently low. In runs with  $\text{PbCl}_2$  as charge material, for instance, a Pb line half-width of about  $1.5$  mm was obtained with the total  $\text{Pb}^+$  current of the order of  $10$   $\mu\text{A}$  (see fig. 3). Very rough estimates of the variation of the minimal line width with ion current indicated an  $I^{1/5}$  dependence in the region  $I < 100$   $\mu\text{A}$  instead of the  $I^{1/2}$  relationship expected from space charge influence (Eq. 10). The explanation mainly lies in the possibility of compensating the mutual repulsion of the ions, acting as a negative lens, by adjusting the electrostatic lens system to shorter focal length. As pointed out by Thulin [12], however, this will enhance the risk of spherical aberrations. Thus an upper limit will be put to the ion currents it is possible to focus in a machine of this type.

After a reconstruction of the ion source (section 3.5), aiming at increased ionization efficiency, the difficulties in handling higher ion currents became more apparent. This may be qualitatively explained by assuming a pronounced "plasma focussing" [27], which was earlier prevented by the relatively long outlet canal. The result is a constriction (waist) with high ion current density outside the ion source and a subsequent larger spread of the beam. In this way the virtual object, idealized as a point source, moves towards the lens system as illustrated by fig. 2a and b, demand-

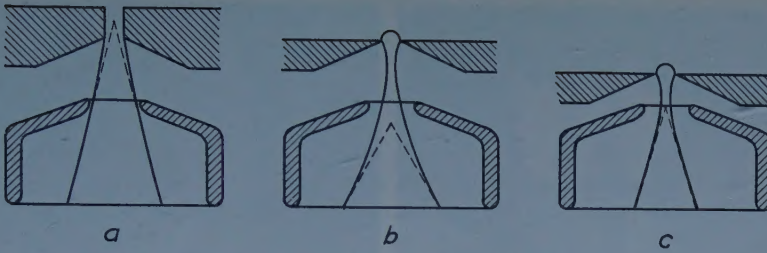


Fig. 2. Various cases of beam extraction from the ion source.

ing a reduced focal distance if the ion beam is still to be rendered parallel. Again aberrations are to be expected.

From experiments with the Stockholm separator, Thulin [12] reached a similar conclusion as Koch did earlier, namely that the line width remained approximately unchanged when the focal plane was moved to a distance from the exit boundary equal to the radius of curvature by letting the lens system form a divergent beam. In this operation the separator is run as a  $90^\circ$  sector field apparatus, though with the axial symmetry of the ion source and the spherical lens system retained. An additional cylinder lens combination for adjusting the heights of the lines and preferably placed at the magnet entrance is then necessary. The improved conditions were ascribed to a considerable reduction of spherical aberrations in the lens system with increased focal distance.

The advantages of the parallel beam focusing in view of the actual applications made a change to a sector magnet arrangement less attractive in our case. In order to extend the region of usability to higher ion currents we have instead considered the possibilities of improving the properties of the beam forming system. Rough estimates indicated that the influence of space charge would be insignificant in the later part of the lens system with the total ion currents (a few hundred  $\mu\text{A}$ ) and final beam diameters (seldom less than 2 cm) desired. Therefore attention was concentrated on the region where the main increase in beam size takes place, that is between the outlet and the first lens gap (suction electrode—1st grounded electrode).

Referring again to fig. 2b, it is obvious that the angle of divergence increases behind the beam constriction. Consequently, in order to reduce the opening angle seen by the lens system, this should be placed as close as possible to the waist, which in addition reduces the width of the beam at the first lens gap (fig. 2c). In this respect the use of a suction electrode is advantageous; its comparatively low potential relative to the ion source diminishes the danger of electrical breakdown. A high efficiency of the ion source works in the same direction [28].

Material problems and different shapes of the outlet plate and suction electrode [cf. 29, 21] have also been studied in this connection. Further it is easily understood that a change of the ion current will move the virtual object point, and thus a modulation of the output from the ion source must be avoided. In order to achieve this the A.C. components of the supplies have been suppressed.

These various measures [30] have led to a considerable improvement of the performance. This is illustrated by fig. 3, which shows the stable Pb spectra at good focusing conditions before and after the reconstruction of the ion source and the acceleration system. It should be added that the overall efficiency of the separation process is at least a factor 10 higher in the later case.

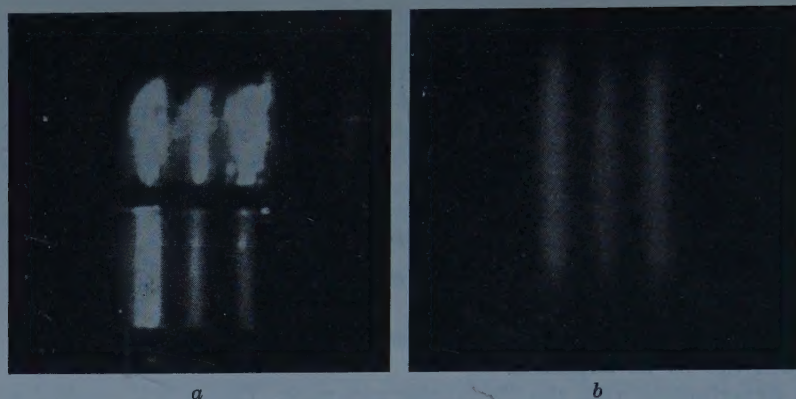


Fig. 3. Photographs of the fluorescent screen showing lines due to stable lead isotopes, *a*, with the original ion source and lens system, *b*, after the reconstruction discussed in section 3.4. The linear scale is given by the distance between neighbouring lines = 3.5 mm. In case *a*, the removal of electrical charges from the screen was insufficient as seen from the upper part of the picture. The wire gauze placed in front of the screen in case *b*, on the other hand caused some blurredness of the lines. The ion current of the three lines together was in both cases of the order of 10–20  $\mu\text{A}$ .

### 3.5. Ion source

The ion source is of the oscillating electron type with an axial magnetic field [31]. It was originally chosen because of some promising results reported by Kistemaker and coworkers [32, 33], indicating in particular its suitability in our case where the ion source magnetic field is independent of the main magnet. In order to restrict the field to the region of ionization, thereby avoiding a feared interference with the ion accelerating system, an iron yoke as described by Veenstra and Milatz [34] was introduced, one of the pole tips simultaneously forming the outlet plate (fig. 4).

As the source was planned right from the start for work on solid charge material a separate heater for a small evaporation oven was incorporated and placed immediately below the anode cylinder. The idea was that vapour from the oven should enter directly into the discharge through a hole in the anode. An opposite hole formed the inlet for gaseous charge material or support gas. By means of a vacuum gate, using a needle type of valve tightening into the outlet canal [cf. 26], the solid charge could be rapidly introduced without breaking the high vacuum in the separator as indicated in fig. 4.

Though this ion source was successfully used in several investigations (see section 4) some drawbacks soon became apparent, the most prominent being

1. The condensation of evaporated material on the well-cooled electrode leads and chamber walls, resulting in low ion yield with solid charge material.
2. The position of the oven close to the discharge-heated anode cylinder, making temperature control of the oven difficult.

This led to a reconstruction, bringing the ion source more in accord with the principles mentioned already in section 2.1 (fig. 5).

The ionization now takes place in a closed chamber, thermally insulated from the surroundings [cf. 35]. It is ordinarily heated from the filament, but the temperature may be further increased by leading current through the spirally wound tungsten

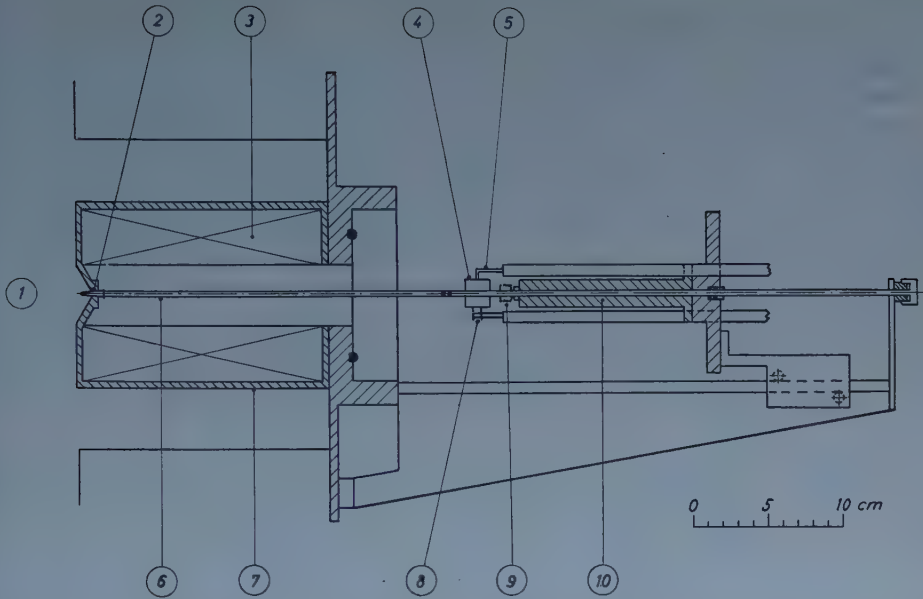


Fig. 4. Axial cross section of the original ion source showing the interior parts removed for charging the oven. 1. High vacuum. 2. Pole piece with outlet opening. 3. Magnet coil. 4. Cylindrical anode. 5. Gas inlet. 6. Needle valve for vacuum gating. 7. Iron yoke. 8. Evaporation oven. 9. Filament. 10. Pole piece.

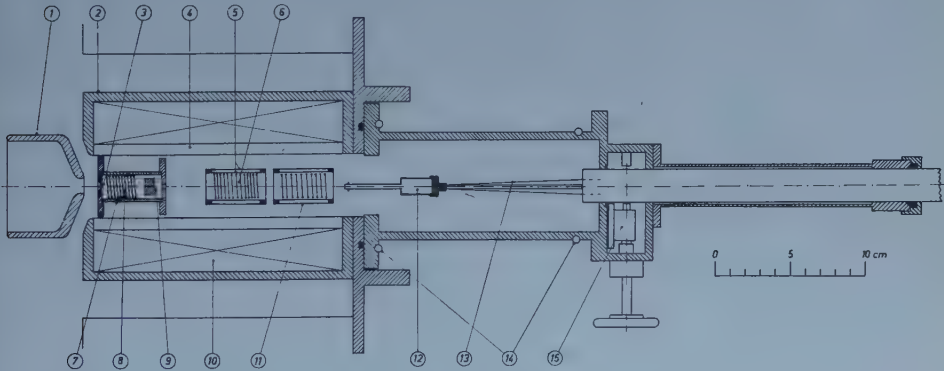


Fig. 5. Axial cross section of the new ion source. (Supports and current leads left out for clarity.) 1. Suction electrode. 2. Iron housing. 3. Outlet opening. 4. Cooling mantle. 5. Radiation shields for oven. 6. Oven heater. 7. Discharge chamber. 8. Directly heated anode. 9. Filament. 10. Magnet coil. 11. Alternative oven for materials of higher vapour pressure. 12. Container for the charge material. Shown removed from its running position inside the heater (6). 13. Thermocouple leads. 14. Cooling pipes. 15. Vacuum gating valve.

anode [cf. 36]. The iron yoke has been left out, giving place to a new oven arrangement. The temperature of the oven is kept at preset values by means of a thermocouple regulator working on the heater current. A more detailed description of the ion source will be given elsewhere [37].

### 3.6. Constructional features

Figure 6 shows a somewhat schematized horizontal section of the mass separator drawn to scale.

As mentioned before, the mean radius of curvature is 100 cm. The second principle parameter determining the size of the magnet, namely the gap height, was chosen as 8 cm. This does not allow the ion collecting device to be placed inside the magnetic field which would be demanded for a strict fulfillment of the focusing principle (section 3.1). Instead the pole pieces have been bevelled as shown in the figure. Because of the stray field effect, which has been corrected for according to Coggeshall [38], the deviation from the ideal case will be insignificant. At the entrance side the pole faces are curved with the radius 100 cm (cf. section 3.1). The pole tips are 22 cm wide, ensuring a usable width of 10 cm, where the field is homogeneous within 1% in radial direction. To achieve this the parallelism of the pole surfaces has in addition been adjusted to about 0.2 mm. The relative mass difference covered is 20%.

The magnet is excited by two series-coupled coils with separate cores. The system of winding the coils is the one described by Coonrod *et al.* [39], that is, sections of copper strips surrounded by water-cooled flanges. A motor-generator delivers the magnet current, which is electronically stabilized to a factor  $> 10000$  and freed from ripple according to principles given by Sommers Jr. *et al.* [40]. The maximal current, 43.5 Amp, corresponds to 6000 Oersted, which is sufficient to deflect the heaviest atomic ions at full acceleration voltage.

The stainless steel cylinders forming the electrostatic lens system are enclosed in an iron housing (to shield for stray fields), the first three of them, as also the ion source, being adjustable relative to the fourth which is fixed to the housing. The lens gaps are about 10 mm wide and the cylinder diameter at the last two gaps is 10 cm, intended to allow a maximal final beam diameter of 4 cm [cf. 8]. Visual control of the lens system during operation is possible through lead-glass windows in one of the chamber walls.

The lenses receive their potentials from a compact, oil insulated high tension set, which in addition contains the ion source supplies. The main rectifier, giving the accelerating voltage  $V_A$  (max. 70 kV), consists of two diode valves and has a filter circuit calculated for a maximal ripple of 25 V at 1 mA load. The focusing potential ( $V_F = 0.10 V_A - 0.95 V_A$ ) is taken from a potential divider across  $V_A$ , while the suction electrode is fed from a selenium rectifier, giving  $V_S = V_A - (V_A - 15 \text{ kV})$ . A common voltage stabilization is arranged by means of a Sorensen regulator on the A.C. side with a stated constancy within  $\pm 0.2\%$ .

The collector housing, which can be shut off from the deflection chamber by means of a vacuum valve, contains a holder usable for different collector arrangements. By means of a shaft through a Wilson seal it can be moved in the longitudinal direction of the spectral lines. A fluorescent screen behind a glass window makes possible visual control during the adjustment of focusing conditions and position of the spectrum. While this is going on the collector is kept below the lines and is slid upwards to operating position when collection can start. The "sniffers" of the line positioner (section 3.3) are movable along the mass spectrum, and a "current collector" on a separate holder is connected to a current integrator [41], allowing simultaneous reading of the ion current, with intervals from  $1 \mu\text{A}$  to  $10^3 \mu\text{A}$  full scale deflection. (In the actual applications the current collector is always set to work on a stable carrier isotope). A typical collector arrangement is sketched in fig. 7.

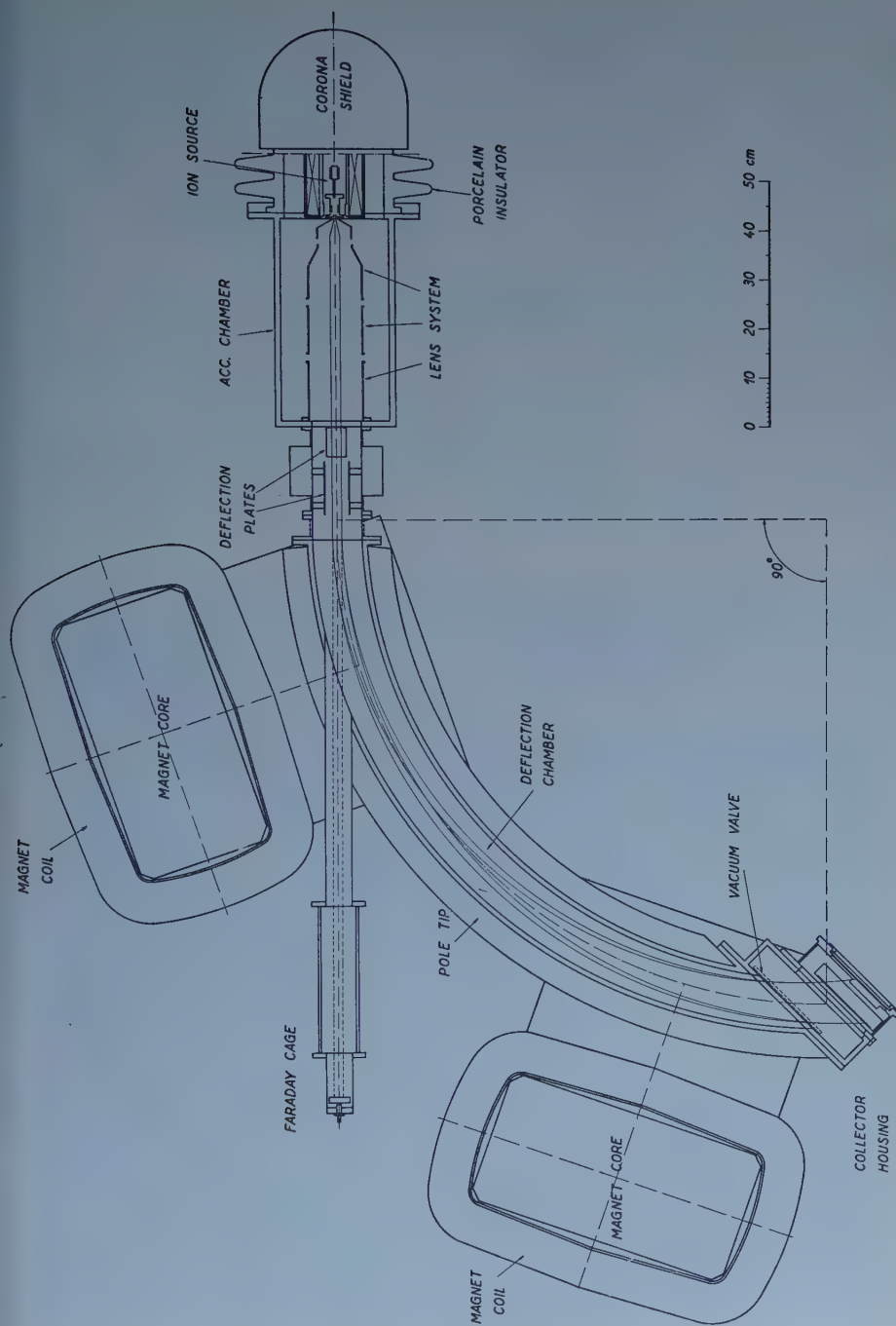


Fig. 6. Principle components of the mass separator.

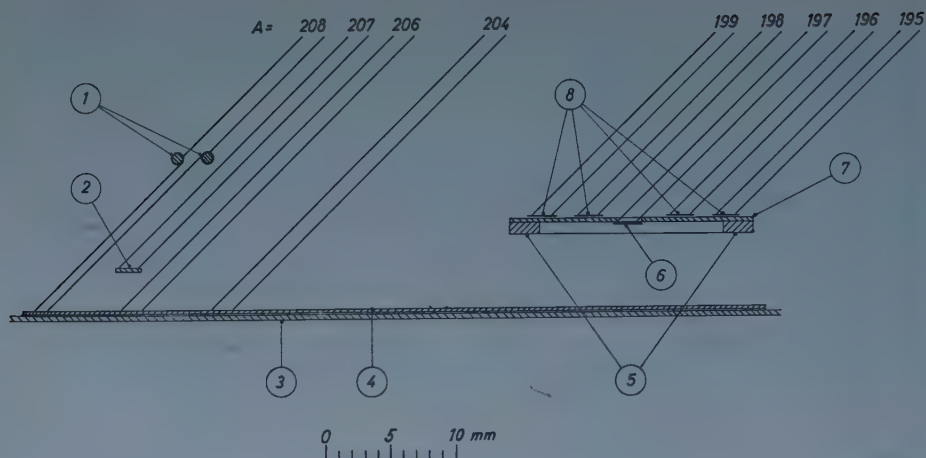


Fig. 7. Collector arrangement for the separation of lead isotopes. 1. "Sniffers" of the line positioner. 2. Collector connected to the current integrator. 3. Fluorescent screen. 4. Metal grid to remove charges from the screen. 5. Sample holder for the  $\beta$ -spectrometer with 6. Backing for the  $\beta$ -spectrometer sample. 7. Plate with defining slit. 8. Additional collector foils.

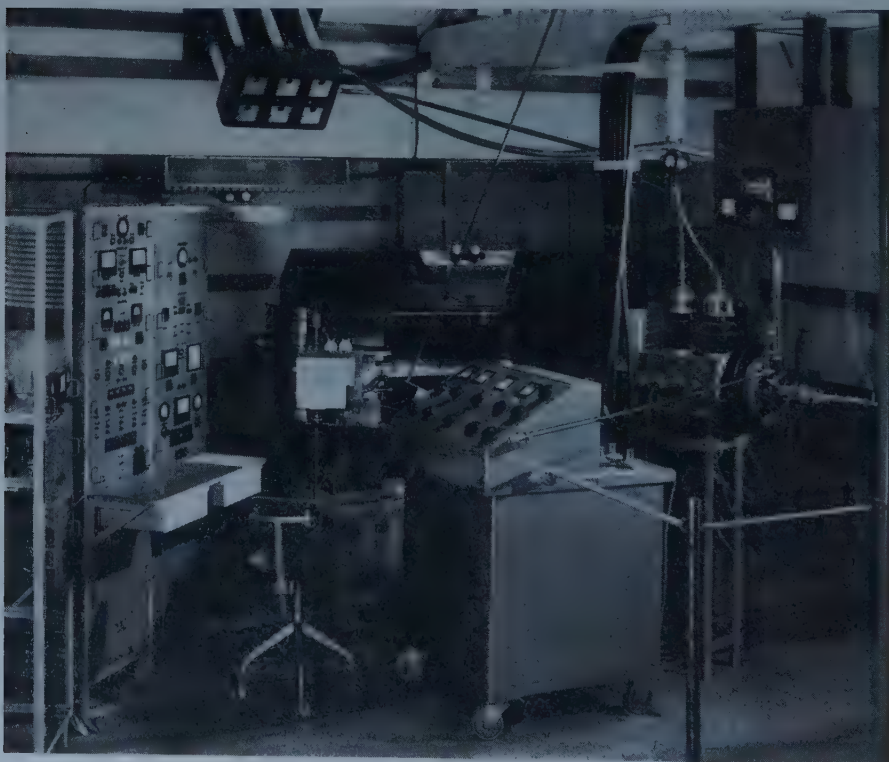


Fig. 8. View of the mass separator. The ion source to the right.

An operating pressure of about  $10^{-5}$  mm Hg is maintained by a 500 l/s (at  $10^{-4}$  mm Hg, unbaffled) oil diffusion pump backed by a mechanical pump. The water-cooled baffle of standard design is expected to decrease the capacity of the high vacuum pump by 25%. A small mechanical pump works separately on the ion source and the collector housing in connection with vacuum gating. Pirani and ionization gauges are used to measure the fore and high vacuum, respectively, and a Philips gauge governs a protection circuit acting on the high tension equipment, the diffusion pump and the ionization gauge.

There are several additional protective measures shutting off appropriate supplies at overloading of the  $V_A$ -rectifier, when opening an enclosure around the ion source part of the separator, at low speed or high temperature of the oil circulated to cool the ion source, and at low water speed in the heat exchanger where the oil is cooled down. In a similar way the magnet coils and the vacuum pumps are protected against failure of the water cooling. Several of the more important voltages and currents, for which there are no separate meters, can be checked during operation by means of a central instrument with a 14-step change-over switch.

A view of the assembled mass separator is shown in fig. 8.

## 4. Applications

### 4.1. Nuclear spectroscopy in the lead region

The mass separator has been frequently used in investigations of highly neutron-deficient isotopes of lead and their decay products [42]. The mass numbers 199 through 195 of lead were obtained by  $Tl(p, xn)$  reactions with proton energies ranging from 70 MeV to 115 MeV. At these energies the excitation curves are far from sharp and conversion electron lines of more than ten different activities were sometimes distinguished in  $\beta$ -spectra of chemically prepared samples. As several of the half-lives were similar the need for mass separation was apparent.

Using mass separated  $\beta$ -spectrometer sources altogether about 30 conversion lines in the decays of 1.5 hr  $Pb^{199}$ , 7.4 hr  $Tl^{199}$ , 2.4 hr  $Pb^{198}$ , 5.3 hr  $Tl^{198}$ , 42 min  $Pb^{197m}$ , and 2.7 hr  $Tl^{197}$  were identified. In the remaining cases the mass assignments of half-lives were confirmed by disintegration curves of mass separated samples. Some complementary information was obtained by scintillation spectrometer measurements of  $\gamma$ -energies. It was sometimes convenient to collect several mass numbers simultaneously on separate strips and take one of them to the  $\beta$ -spectrometer, another to the  $\gamma$ -spectrometer, and others to ordinary G.M. counting.

As to the separation technique,  $PbCl_2$  was found to be a suitable charge material. A serious limitation, however, lay in the efficiency of the process when the original ion source was used. Considerations of the amounts it is possible to produce in the synchro-cyclotron and those needed for the  $\beta$ -spectrometer indicated a desirable overall efficiency of 10% [cf. 14]. Normally less than 1% was obtained. This was one of the main reasons for reconstructing the ion source (section 3.5).

With the improved running conditions now existing the investigations have been taken up again [43]. Fig. 9 shows a section of an automatically recorded conversion electron spectrum of 37 min  $Pb^{196}$ , taken with the double-focusing  $\beta$ -spectrometer. For comparison the spectrum of an unseparated sample obtained under identical bombarding conditions has been included. It should be especially noticed that even the strongest line complex in this spectrum which belongs to the decay of 42 min

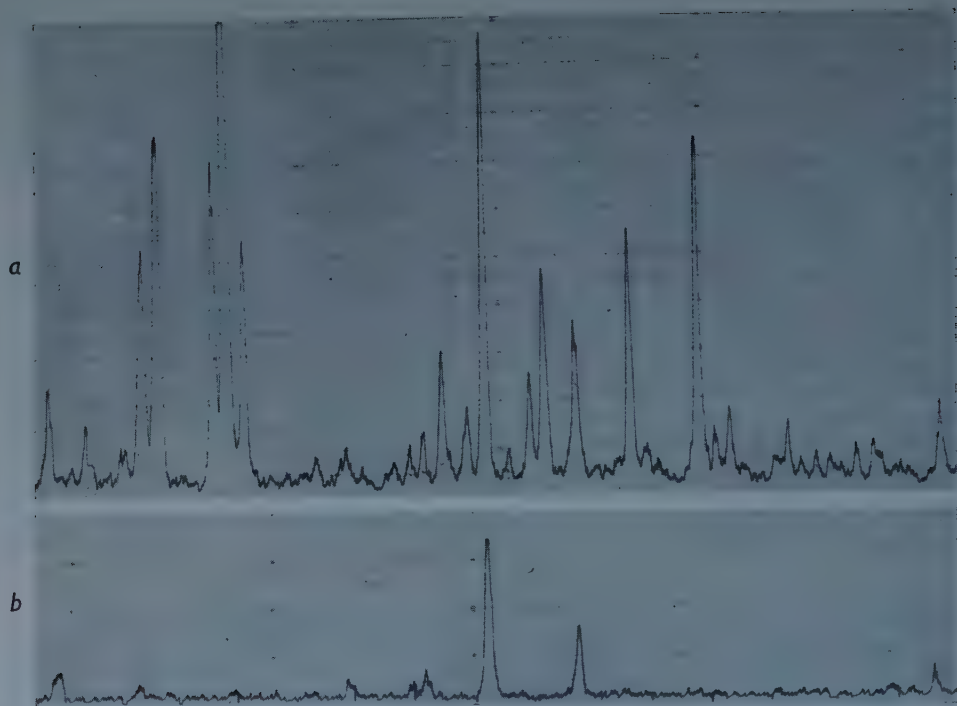


Fig. 9. Automatically recorded conversion electron spectra in the energy region 100–250 keV of lead samples from the irradiation of natural thallium with 105 MeV protons, *a*, after chemical separation and vacuum evaporation, *b*, after chemical separation and mass separation with collection of  $A = 196$ . Sensitivity in both cases: full scale deflection = 100 counts/sec.

$\text{Pb}^{197m}$  is absent in the mass 196 spectrum. This shows the high isotopic purity of the separated sample.

Using the same sample the  $K$  and  $L$  lines of a 426 keV transition following the decay of 1.8 hr  $\text{Tl}^{196}$  were studied by point to point plotting at about  $3^\circ_{00}$  spectrometer resolution. With regard to the solid angle and G.M. slit used this resolution indicates that the effective width of the sample was in fact less than 2 mm, which was the width of the defining slit during the mass separation. The total  $\text{Pb}^+$  current amounted to about  $50 \mu\text{A}$ .

A recent application of considerable interest concerns the measurement of half-lives in the  $\mu\text{s}$ -ms region. If, for instance, the metastable state is fed by electron capture, the accompanying X-ray can be used to start a time analyser set to work on some  $\gamma$ -transition originating in the metastable state. When several isotopes of the parent element are present in the sample, X-rays from all of them can trigger the analyser, bringing about a "background" which may overshadow the decay looked for. This situation was encountered in attempts to study the 460 keV E3 transition in  $\text{Tl}^{202}$ , appearing in the decay of 3.6 hr  $\text{Pb}^{202m}$  [44]. With a mass separated sample Tove succeeded in determining the E3 half-life as  $475 \mu\text{s}$  [45]. It should be especially noted that the sample was much too strong for this measurement; only a fraction of it could be used. Another part was studied in Stockholm by a different method, confirming the result within the limits of error [46].

A similar technique was employed in the case of a 48 keV E2 transition in  $\text{Hg}^{198}$ , fed from 1.8 hr  $\text{Tl}^{198m}$  [47]. Because of the high conversion coefficient the time analysis was made on the  $L$  electrons, detected by a thin plastic scintillator [cf. 48]. There was, however, no indication of a half-life in the region  $100\ \mu\text{s}$ – $1\ \mu\text{s}$ , which was further extended downwards to  $50\ \text{m}\mu\text{s}$  by a delayed coincidence technique [49].  $\text{Tl}^{198m}$  was in these experiments produced by irradiation of  $\text{HgCl}$  and mass separated with  $\text{TlCl}$  as the charge material.

#### 4.2. Spallation yields

It should first be pointed out that the mass separation method can by no means replace ordinary radiochemical techniques in spallation and fission studies. Owing to the different ionization probabilities of the elements it will in general be possible to determine only relative yields of isotopes of the same element. This requires further that mass discriminating effects can be neglected, that the collection efficiency is the same for all mass numbers concerned, and that the deposits have the same dimensions and intensity distributions. Under these presumptions mass separation can be used as a valuable complement to the chemical methods.

That in some cases, notably in the medium low mass region, information can be gained independent of chemistry was shown, however, in an investigation where metallic vanadium was bombarded with 187 MeV protons [50]. The target was immediately transferred to the ion source of the separator and in this way the separation could be started about ten minutes after the irradiation. Stable argon was used as support gas for the discharge and also to establish the mass scale. The evaporation of radioactive atoms from the vanadium target placed close to the discharge (in the old ion source) was wholly uncontrolled. Mass numbers 41–48 were simply collected during 15-minute runs and their disintegration subsequently followed in a G.M. counter. The decay curves obtained were relatively uncomplicated, containing three components at the most. Altogether eleven isotopes of A, K, Sc, Ti, and V with half-lives between 22 min and 3.3 d were identified. The relative yields calculated for K and Sc isotopes gave additional data for comparison with an empirical cross section formula put forward by Rudstam [51].

A more "normal" use was made of the mass separator in a study of gallium isotopes from spallation of arsenic [52]. The need for mass separation in this investigation arose from similarities in half-life,  $\text{Ga}^{66}$  (9.6 hr),  $\text{Ga}^{72}$  (14 hr), and  $\text{Ga}^{73}$  (4.8 hr) forming one group,  $\text{Ga}^{68}$  (68 min) and  $\text{Ga}^{70}$  (22 min) another.  $\text{GaF}_3$  was found to be a suitable end product of the chemical separation and as suitable a charge material for the new ion source. Estimated separation efficiencies for gallium were of the order of 10%, and stable ion currents of more than  $100\ \mu\text{A}$  of  $\text{Ga}^{69}$  were satisfactorily focused.

#### 4.3. High energy fission yields

A very good example of the usability of the mass separator in yield measurements was furnished by the iodine isotopes formed in fission of uranium induced by 170 MeV protons [53]. After chemical separation the iodine fraction contained appreciable amounts of about ten radioactive isotopes with half-lives ranging from 25 minutes to 60 days. Ordinary G.M. counting techniques were certainly not able to distinguish the components. On the other hand this was highly desirable, not least because of the presence of four shielded nuclei, that is nuclei which can only be formed as primary reaction products. After mass separation the relative yields of eight isotopes

could be determined. In this way valuable information about the charge distribution in high energy fission was obtained [54].

For reasons of rapid chemistry AgI was chosen as the charge material. Because of insufficient temperature control of the evaporation oven (the old ion source was used) it was found necessary at least to start the discharge with support gas (argon). The separation conditions were not ideal, which was also revealed by careful analysis of the various decay curves. Some of the findings are of general interest and will be briefly reviewed, though a more detailed discussion is given in the communication previously referred to [53].

The width of the mass lines was normally kept below 3 mm as visible on the fluorescent screen (very approximately equal to the half-width), the dispersion being 5 mm. Nevertheless up to 5% of an activity assigned the mass number  $A$  was sometimes found on the  $A-1$  collector and as much as 2% on the  $A-2$  collector. This may be ascribed partly to electrical breakdowns occurring frequently in the beginning of the separations and partly to gas scattering, enhanced by the use of support gas.

A higher percentage showed up as a contamination of mass number  $A + 1$  because of hydride ion formation. The hydride content was found to decrease during the runs, giving an average of 10–15%. This decrease probably was the cause of a curious behaviour of the line positioner noticed when the "sniffers" set to operate on  $A = 127$  were 1 mm wide and spaced 2 mm, allowing one of them to be reached by the "tail" of the hydride line of  $A = 128$ . Then the equilibrium of the stabilizer was displaced with the change in intensity ratio  $A = 127/A = 128$ , resulting in a side-moving of the whole mass spectrum. This emphasizes that care must be taken in handling this type of stabilizer.

In one of the runs it was possible, by activity comparison with the chemically isolated  $\text{Sr}^{89}$ , whose high energy fission cross section was known [55], to estimate the separation yield of iodine as about 0.2%. Simultaneously, however, 5–10% efficiency for xenon formed in iodine decay was indicated.

#### 4.4. "Hot atom chemistry"

Though it falls somewhat outside the subject of this communication an interesting application proposed by G. Giacomello, Rome, and U. Croatto, Padua, will be briefly mentioned.

The basic idea is that ions accelerated to some tens of keV may use their kinetic energy to cause chemical reactions while being trapped in an inorganic or organic target material. This corresponds to a kind of controllable hot atom chemistry. Because of the small penetration power of the ions the reactions will take place within a very limited thickness of the target. An analysis of the yields of different reaction products is feasible only if ions of a radionuclide is used. In order to obtain these carrierfree and thus to avoid sputtering action and excessive heating of the target it is convenient to perform a mass separation simultaneous to the acceleration of the ions.

A number of runs were made in the mass separator, using  $\text{C}^{14}$  ions and 30 kV acceleration voltage [56]. The carbon was introduced as  $\text{CO}_2$  and, as expected, the formation of  $\text{CO}^+$  was favoured in the ion source. It was possible, however, by raising the anode potential, to increase the  $\text{C}^+$  content to almost half that of  $\text{CO}^+$ . The stable  $\text{C}^+$  current used was of the order of 10–20  $\mu\text{A}$  and the yield of  $\text{C}^+$  as a fraction of the total amount of carbon added was in a preparatory run estimated as about 0.25%. The results obtained [57, 58] were promising and the investigations will be continued with a mass separator being built in Rome.

## 5. Conclusions

To sum up the present situation, the mass separator has reached a stage of development permitting in general a rapid and fairly complete separation even of heavy mass isotopes at a total ion current of a few hundred  $\mu\text{A}$ . The most favourable conditions, resulting in overall separation efficiencies of the order of 10%, are obtained when the charge material is a compound (or an element) whose vapour pressure relations require oven temperatures in the interval  $300^{\circ}\text{C}$ – $800^{\circ}\text{C}$ . Then the ion source can as a rule be run without any support gas, which is an advantage in view of the relatively low pumping speed. (It would be desirable for the work on gaseous charge material to have this raised to, say, 1500 l/s.)

It should be pointed out, however, that the use of quite different separation techniques may sometimes lead to superior results. The vanadium experiments (section 4.2) indicate the interesting possibility of replacing a chemical separation by a kind of fractionated distillation of low-melting elements from a refractory target material in the evaporation oven. This would be of a special importance in the rare earth region, where in fact neighbouring elements in some cases show widely differing vapour pressure curves [59]. Similarly noble gases can be released from, for instance, a bromide or an iodide by heating the compound in an external container, connected through a cold trap to the gas inlet.

Very probably the last word has not yet been said as to ion source construction. There are indications [28] that it may be possible to raise the ionization efficiency considerably in many cases. A promising technique which remains to be tested in the present apparatus is offered by the sputtering ion source [60]. This is especially suitable for extremely low vapour pressure material.

It has long been realised that fluorescent screens are not ideal in controlling the sharpness and position of the lines. The pictures obtained depend on the intensity of the lines and the condition of the screen. In particular it is disturbing that the fluorescence material becomes worn out during a run, making continuous control difficult. Thus a mechanical beam analyser recently introduced at the Copenhagen separator by Nielsen and Skilbreid [61] represents a considerable improvement. It works with a thin measuring probe sweeping across a part of the mass spectrum and reproduces the intensity distribution on an oscilloscope screen. Space problems unfortunately prevent the incorporation of a similar device with our separator. On the whole a more spacious collector housing would be advantageous.

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### G. ANDERSSON, *Preparation of radioactive samples*

Later development work has been carried out by Dipl. phys. Jan Olof Burgman, who also performed several of the separations mentioned in section 4 of this communication, and Ing. Rolf Ringh. Fil. kand. Bo Jung has contributed discussions about ion extraction and beam formation.

The extraordinarily good cooperation with the other separator groups in Sweden and Denmark, manifested in frequent communication of ideas and results, has greatly promoted the work. In particular I am indebted to Dr. Ingmar Bergström and Dr. Sigvard Thulin, who introduced me into the technique of electromagnetic isotope separation during a two months' stay at the Nobel Institute in 1949.

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